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14. ABSTRACT								
Single-frequency did	ode lasers were stabil	ized to 500Hz precision	on at 1.5 microns and	to 20Hz at 793	nm on 10-ms time scales using	narrow		
spectral holes in abs	orption lines of Er as	nd Tm doped cryogen	ic crystals as frequenc	y references an	d without vibrational isolation	of either		
the laser or frequence	v reference. Kiloher	tz stability for 100 sec	conds is provided by t	hese techniques	s, and that performance is exter	ndable to		
longer time scales w	ith further developm	ent. Miniaturized lase	ers and 2mm-sized ref	ference crystals	will provide compact portable p	packages		
with a closed cycle of	ryocooler. The achie	eved frequency stabili	zation provides ideal	lasers for high-	resolution spectroscopy, real tit	me		
optical signal proces	sing based on spectr	al holography, and oth	her applications requi	ring ultra-narro	ow-band light sources or cohere	ent		
detection. Feedback	for stabilizing the fr	requency of external c	avity diode lasers was	derived from F	Pound-Drever-Hall frequency			
modulation probing	of the spectral hole a	ind used to control did	ode laser current and	grating tilt. Sta	bilization to spectral holes is ap	plicable		
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to DFB lasers and other lasers and to other transducers like acousto-optic or electro-optic frequency shifters. Studies of frequency reference materials provided crystals that are programmable long-term references with spectral holes persisting up beyond liquid nitrogen								
temperatures. Crystal frequency references are insensitive to vibration and could be mass-produced.								
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Final Technical Report

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AFOSR-DEPSCoR Contract Number F49620-96-1-0466

VERY-NARROW-LINE SEMICONDUCTOR LASER AND OPTICAL CLOCKS BASED ON SPECTRAL HOLE BURNING FREQUENCY STANDARDS

submitted by

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Objectives

The objectives throughout remained as stated in the proposal: to develop highly stable frequency references at selectable wavelengths by locking diode lasers to persistent spectral holes in crystals, to develop new hole burning materials as spectral hole frequency references, and to develop stabilized tunable lasers and frequency locking techniques.

The resulting optical frequency references differ from atomic clocks and frequency standards in several ways: the frequency is selectable from anywhere within the inhomogeneously broadened absorption of the reference crystal rather than restricted to the line center of an atomic transition, it is optical rather than a microwave reference, and the medium is a small millimeter scale piece of cryogenic crystal rather than an atomic beam, laser cooled atoms in a trap, or atomic fountain.

Summary of Results

The original goals have been realized. The major results are:

- 1. We have reported the first experimental demonstrations of using spectral holes as frequency references for stabilizing laser frequency. We first demonstrated locking diode lasers to persistent spectral holes in Tm:CaF₂:D at 798 nm. [Opt. Lett. **24**, 1038-1040 (1999)]
- 2. We developed a new persistent spectral hole burning material for 798 nm, Tm:CaF₂:D⁻, and the spectral holes persist to over 60 K. [Phys. Rev. B. **59**, 14328-14335 (1999)]
- 3. We worked on a similar material for operation at the *important communication band of 1.5 microns*. [To be submitted to Phys. Rev. B]
- 4. In addition we have developed another new technique of *locking diode lasers to regenerated* transient spectral holes. This was first demonstrated using Tm³⁺:YAG (Tm³⁺:Y₃Al₅O₁₂) at 793 nm. [Phys. Rev. B **62**, 1473-1476 (2000)]
- 5. We demonstrated the first locking of diode lasers to spectral holes at the important communication band of 1.5 microns. This used Er³⁺:Y₂SiO₅ as the spectral hole burning material. [To be submitted to Phys. Rev. B]
- 6. We have developed 20 Hz-level stability out of 4 x 10¹⁴ Hz at 793 nm diode laser wavelengths and 200 Hz-level stability out of 2 x 10¹⁴ Hz at 1.5 micron diode laser wavelengths. A number of spectral hole burning materials have characterized for operation in each of these spectral regions.
- 7. The application of these stabilized sources to real time optical signal processing based on spectral hole burning and coherent optical transients has enabled significant progress in the optical signal processing area by other groups at Montana State University. A particularly exciting aspect of this is that we showed the spectral hole burning materials that are being used as optical signal processing materials may be used as the frequency references, too.
- 8. In addition, we showed that these new light sources also provide exceptional advancements in the sensitivity of coherent transient spectroscopy, allowing measurements to be carried out on solids with world record precision.

Accomplishments/New Findings

Programmable laser stabilization using persistent spectral hole burning Opt. Lett. 24, 1038-1040 (1999)

We reported the first demonstration of laser frequency stabilization to persistent spectral holes in a solid-state material. The frequency reference material was deuterated $CaF_2:Tm^{3+}$, prepared with 25 MHz wide persistent spectral holes on the ${}^3H_6 \rightarrow {}^3H_4$ transition at 798 nm. The beat frequency between two lasers independently locked to persistent spectral holes in separate crystal samples showed frequency precision of 780 \pm 120 Hz for 20 ms to 50 ms integration times (based on measured root Allan variance).

Unlike absolute frequency gas phase transitions or the periodic spacing of highly stabilized Fabry-Perot cavity resonances, spectral holes can be prepared at any frequency within a broad inhomogeneous absorption profile (15 GHz in the material demonstrated here). Such a frequency reference could be used as a programmable and transportable secondary frequency standard. Highly frequency-stabilized laser sources are also desirable for improving carrier frequency and phase stability for applications such as time-domain and frequency-domain optical storage and erasure, optical processing, network routing, wavelength division multiplexing, long baseline interferometry, ultra-high resolution spectroscopy, or Doppler lidar velocimetry.

Several lasers can be stabilized to multiple spectral holes, either in the same or separate absorption bands, with arbitrary frequency separations. The customizable beat frequency between two or more stabilized lasers may also be used to synchronize high speed optical computing operations and data transfer and to provide clocks for other applications.

The spectral hole burning material used in this demonstration was deuterated CaF₂:Tm³⁺. The nominal Tm³⁺ concentration was 0.05% (molar) and the linear absorption was ~60% at line center. Many crystallographically inequivalent sites exist for the Tm³⁺ ions in this system, and in our material studies (discussed below) the L1 site has been shown to exhibit persistent spectral hole burning with hole widths of 25 MHz on the $^3H_6 \rightarrow ^3H_4$ transition of Tm³⁺ at 798 nm. The hole burning mechanism involves localized migration of deuteride (D') ions with a barrier activation energy of 1800 cm⁻¹. The holes have been shown to be persistent without measurable degradation for at least 48 hours at 1.7 K and to survive thermal cycling to 60 K. Persistent spectral holes can be created at higher temperatures with some increase in linewidth. A sample temperature of 20 K gives hole widths of 150 MHz, which would give a projected frequency stability of a few kHz. Commercial closed-cycle refrigerators can readily chill below 10 K making this operation practical without cryogenic fluids. Operation at liquid nitrogen temperature is not practical with this material, but it may be with others.

The setup shown in Fig. 1 contained two GaAlAs external cavity diode lasers (Littman-Metcalf configuration). The lasers were externally modulated with electro-optic phase modulators (EOM) at 23 and 25 MHz respectively for frequency locking to the spectral holes with the Pound-Drever-Hall technique. This method used frequency modulation (FM) spectroscopy of the spectral hole transmission to provide a corrective feedback signal to a servo amplifier, which rapidly modulated the laser current and slowly adjusted a piezo-electrically driven grating for optical feedback to keep the current servo within operating limits.

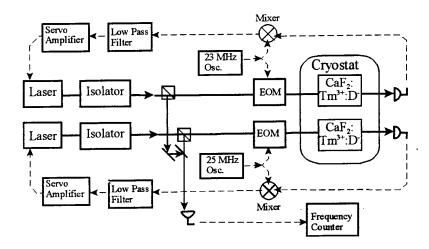


Fig. 1. Experimental setup for frequency locking to spectral holes, and beat frequency characterization of relative laser stability.

Each laser was independently locked to a spectral hole in a separate crystal. A single cryostat held both crystals immersed in superfluid helium at 1.9 K to improve thermal equilibration and temperature stability. The stability of the frequency difference between the two lasers was measured by heterodyne detection and monitored on a frequency counter. The frequency of the heterodyne beat signal could be chosen arbitrarily by the relative frequencies of the two spectral holes.

The locking stability depended on the laser power through two opposing factors. Low irradiance (power per unit area) in the crystal minimized continued hole burning that would modify the reference spectral hole, while high total power on the detector maximized the signal to noise ratio of the feedback loop. In addition, frequency drifts can arise from unintentionally locking slightly off the center of the hole. Ideally, the lock point should remain at the hole center, but in the present implementation the lock frequency was displaced by FM signal contributions from the inhomogeneous line and thermally dependent amplifier offsets at the mV level.

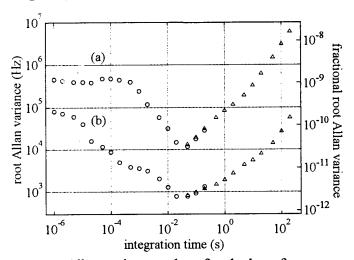


Fig. 2. Root Allan variance values for the beat frequency between two lasers. (a) Lasers free-running, and (b) lasers actively locked to spectral holes in separate crystals. Circles, mean of five 100-sample Allan variances measured directly on the frequency counter; triangles, values calculated from beat frequency data (cf. Fig. 3(a)). Typical uncertainties are $\pm 10\%$, similar to the data point size.

The stability of the beat frequency over different time scales is represented by the Allan variances plotted in Fig. 2(a) for the free-running lasers and Fig. 2(b) for lasers actively stabilized to spectral holes. Typical root Allan variances of 780±120 Hz were measured over 20 ms to 50 ms time scales; quiet periods displayed variances as low as 540±90 Hz over 50 ms. These data demonstrated a stabilization of both lasers to 2 parts in 10⁵ of the 25 MHz hole, or 1 part in 10¹² of the optical frequency.

Persistent Spectral Hole Burning Materials

Persistent spectral hole burning materials at wavelengths accessible to diode lasers, particularly around 800 nm and 1.5 microns were developed and characterized. High resolution measurements of optical properties were carried out over temperature ranges from liquid helium temperatures to 100 K. Several of the materials described below are promising for stabilization using spectral hole burning at elevated temperatures that allow practical use of compact refrigerators.

Persistent spectral hole burning in deuterated CaF₂:Tm³⁺

Phys. Rev. B. 59, 14328-14335 (1999)

Spectral hole burning in the deuteride (D) modified Tm^{3+} centers in CaF_2 was explored. The hole burning mechanism involved localized displacement of the D ions. Two main families of Tm^{3+} - D centers were present; the Li centers yield spectral holes that we measured to be fully persistent for 48 hours at liquid-helium temperatures, while spectral holes of the Mi centers have a hole-recovery time constant of approximately 20-30 s. Hole widths vary from 18–40 MHz (full width at half maximum) for the different centers. Hole burning dynamics were in agreement with a simple and general model that takes into account the finite homogeneous linewidth. The area of the spectral holes was found to be conserved after the sample temperature was cycled up to 70 K.

Almost all previously reported cases of persistent spectral hole burning in rare-earth doped crystals involve photoionization of the rare earth as the hole burning mechanism, limiting such observances to specific ions and hence specific wavelengths. The hole burning mechanism that applies to rare-earth transitions in deuterated CaF_2 involves D^- ion displacement. This mechanism is host specific, rather than dopant-ion specific, and it therefore provides greater wavelength versatility by allowing the relatively free choice of rare-earth dopant.

Our study of persistent spectral hole burning focused on the ${}^3H_6 \rightarrow {}^3H_4$ transition of D compensated Tm³⁺ centers. This transition is of particular interest for applications since it lies near 800 nm in a wavelength region conveniently accessible to cheap and compact GaAlAs diode lasers. No other Tm³⁺-doped crystal, to our knowledge, exhibits persistent spectral hole burning. Indeed other Tm³⁺ systems typically exhibit millisecond time scale hole lifetimes.

Three main single Tm^{3+} ion centers are found in undeuterated $CaF_2:Tm^{3+}$. The principal center is the $C_{4\nu}$ -symmetry A center on which the Tm^{3+} ion is charge compensated by an F^- ion in the nearest-neighbor interstitial position in the (001) direction. The secondary B center with $C_{3\nu}$ symmetry was initially proposed to have its charge compensating F^- ion in the next-nearest-neighbor position in the (111) direction, but more complex configurations have since been suggested to account for the observed dielectric relaxation and anomalous crystal-field splittings. The A and B centers undergo millisecond time scale transient hole burning from population storage in the metastable 3F_4 level. For the cubic center with remote charge compensation, electric-dipole transitions such as the ${}^3H_6 \rightarrow {}^3H_4$ transition are forbidden by inversion-symmetry selection rules.

The introduction of negative D^- ions produces a series of new rare-earth centers that involve D^- ions substituting for interstitial or lattice F^- ions in the vicinity of the rare-earth ion. These centers give rise to a series of additional sharp absorption lines and can therefore be probed selectively using a tunable laser. The rare-earth ion is charge compensated by an interstitial D^- ion residing at

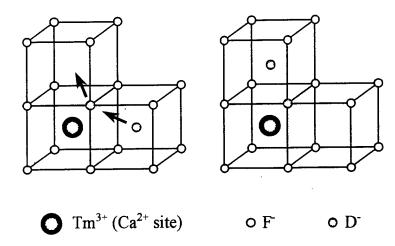


Fig. 3. Proposed site configuration of the L1 spectroscopic center, and the D ion displacement mechanism responsible for persistent spectral hole burning of this center; (a) the original configuration, and (b) the photoproduct configuration.

the nearest-neighbor interstice, in the (100) direction. The C_{4v} symmetry of this configuration can then be reduced by the substitution of further D ions for the F ions of the lattice. The simplest of these low-symmetry centers, having one substitutional D ion in addition to the interstitial, is represented in Fig. 3. For heavier rare earths, such as Er³⁺ and Tm3⁺, a second family of centers appears. Those are most likely based on the $C_{3\nu}$ symmetry configuration of the B center. For the Tm³⁺ centers that we studied, the first group of D-compensated centers derived from the A center are labeled Li (i 50,1,2,3)22 where L0 is the C_{4v} symmetry center with only one D ion. The second group of D'-compensated centers,

presumed to derive from the B center, are labeled Mi.

The absorption transitions of the low-symmetry multiple-D compensated rare-earth centers can be bleached by pumping with a tunable laser. The bleaching behavior of the Li centers involves local displacement of the D ions in the vicinity of the rare-earth ion. From the reduced fluorescence lifetime of these centers, it is known that the principal relaxation pathway for the photo-excited rare-earth ion is to transfer its energy to multiple high-energy (700 cm⁻¹) local-mode vibrations of adjacent D ions. These excited D ions then have some chance of migrating into nearby interstices. The stepwise process illustrated in Fig. 3 illustrates the necessity of a substitutional D ion, the substitutional D ion moves into a vacant interstitial position, and the interstitial D ion takes its place at the substitutional site. This mechanism is supported by the absence of any bleaching behavior for the C_{4v} center involving just a single D ion for any of the rare earths studied to date, showing that a direct tunneling process from one interstitial site to another does not occur at low temperature.

In the spectroscopy experiments, the ${}^3H_6 \rightarrow {}^3H_4$ absorption lines of CaF₂:Tm³⁺:D⁻ were studied. The absorption transitions were clearly divisible into groups of transitions corresponding to the two types of centers. The higher energy group near 12 530 cm⁻¹ is due to the Li centers, with only one line per center, and those lines exhibit the expected reversible polarized bleaching behavior under broadband pumping conditions. The second group of lines near 12 440 cm⁻¹ corresponds to the Mi centers, and these have one or two lines per center, since the corresponding line of the B center is known to be a singlet-doublet transition.

Four of the absorption lines gave long-term or medium-term persistent spectral hole burning. The centers giving rise to these lines are labeled L1, L2, M1, and M4. An estimate of the concentration of each center was obtained by assuming the entire concentration of Tm³⁺ ions in

the crystal to be 0.01% and by taking the oscillator strength of each transition to be the same. This last assumption is a weak one when making comparisons between the two families of centers, but it provided the only simple means of estimating individual center concentrations.

Characterization of the spectral hole burning was carried out using a tunable Coherent 899-21 single-frequency Ti:sapphire laser pumped by a Coherent Sabre argon-ion laser and intensity-stabilized. The spectral holes were measured in transmission at laser intensities reduced by 1 to 2 orders of magnitude relative to the burn intensity. Sample temperatures were controlled anywhere between 1.5 and 300 K in an Oxford Instruments cryostat and were determined using a carbon-glass resistance thermometer.

Of the four absorption lines that exhibit hole burning, two are from Li-type centers and two from Mi-type centers have qualitatively different persistence behaviors. Results are summarized in Table I. The Li-center holes at 798 nm did not show any measurable change over 48 h, the longest period measured, while the Mi-center holes have decay constants of less than a minute. Typically, a laser intensity of 1 mW/cm² was used for burning, and this was reduced by one to two orders of magnitude for the scan cycle. In this regime, shallow holes took tens of seconds to burn, while burning during the scan cycle was minimized. A relatively large laser spot size of 2-3 mm was used to provide sufficient total power for efficient detection.

TABLE I. Characteristic spectral hole burning parameters of the four centers which were found to give long-term spectral hole burning.

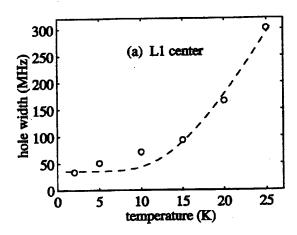
Center	Hole FWHM (MHz)	Quantum efficiency (x 10 ⁻⁴)	Recovery time (s)
Ll	18	8	$> 10^{6} a$
L2	30	20	> 10 ⁶ a
M1	20	300	23
M4	40	100	23

^a Lower limit for persistence dependent on measurement duration

The measured hole widths are much broader than those that have been measured in the transient regime in undeuterated CaF₂:Tm³⁺ or those associated with this transition of the Tm³⁺ ion in other ionic crystals, but they are similar to those observed in CaF₂:Pr³⁺:D⁻. It is possible that spectral diffusion, due to D-motion at one Tm³⁺ site affecting the resonance frequency at another Tm³⁺ site, is responsible for the width.

No decay of the hole amplitude or area was seen for the L1 center for up to 48 h after burning. The persistence of spectral holes in the L2 center was tested for 7 h, with a similar lack of degradation. In contrast, spectral holes burned in the M1 and M4 absorption lines decayed with time constants of 2-3 s with the sample in the dark at 1.7 K.

The temperature dependence of the hole burning process was investigated up to 30 K. Holes were burned and read at a series of temperatures from 2-30 K, with the laser fluence for hole burning fixed at 6 mW/cm² for 1 min. The widths (FWHM) of the resulting holes as a function of temperature are shown in Fig. 4(a), and the hole depth is plotted against width in Fig. 4(b). As expected, holes burned at higher temperatures are broader, due to phonon-induced dephasing, and are therefore shallower. The solid curve in graph (b) represents calculated hole depth and width. The measured decrease in hole depth matches closely the expected behavior arising from homogeneous line broadening. The product of width and depth approximates the hole strength, and for fixed burn parameters this increases with temperature.



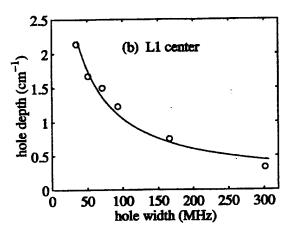


Fig. 4. Temperature dependence of L1-center spectral hole burning. Spectral holes were burned and read with the sample maintained at the stated temperatures. (a) hole width increase with temperature and (b) hole depth dependence on hole width. The solid line in (b) is from simulation, where the hole widths were taken as input parameters.

An important hole burning parameter for device applications is recoverability of the hole after cycling to elevated temperatures. Holes were burned with the sample at 2 K, then the sample temperature was raised to specific intermediate temperature values, held there for 10 min, then recooled to 2 K before being remeasured. Some broadening of the holes with cycling was observed, but the area of the hole was conserved after cycling up to at least 70 K. This broadening most likely arises from thermal relaxation of the D and CaF2 lattice ion positions. These results show that thermally induced relocation of the D ions is inhibited up to at least 70 K, an important result for situations in which occasional sample heating may occur.

We placed a lower limit on the barrier height W for the D^- ion relocalization of the L1 center by considering the thermal barrier hopping rate R to be given by the Arrhenius relation. For an attempt frequency estimated to be $\Omega_0 \approx 2 \times 10^{13}$ Hz corresponding to the ~ 700 cm⁻¹ energy of the D^- vibrational modes, we estimated a barrier height of W = 1800 cm⁻¹.

The hole burning quantum efficiency in the L1 and L2 centers is ~10⁻³. For applications requiring hole stability during long reading cycles, such as laser frequency references, this low efficiency is desirable. The L1 and L2 centers are robust under temperature cycling to at least 70 K, with the conserved hole areas demonstrating that reverse processes are not significant up to such temperatures.

The potential for gated spectral hole burning in this material was explored.

Persistent spectral hole burning in deuterated CaF₂:Er³⁺

To be submitted to Phys. Rev. B

Persistent spectral hole burning has also been studied at 1523 nm in two D modified Er^{3+} centers in CaF_2 . The $^4I_{15/2} \rightarrow ^4I_{13/2}$ transition of the Er^{3+} ion lies in the technologically important optical communications band. We believe this to be the first report of persistent spectral hole burning in the 1.5 μ m wavelength region and also the first report of persistent spectral hole burning for the Er^{3+} ion.

The hole burning mechanism in deuterated CaF₂:Er³⁺ involves localized displacement of the D

ions in the vicinity of the Er³+ ion, as was the case for the Tm³+ material described above. The spectral holes were observed to undergo no measurable change (to be fully persistent) for 48 hours at liquid helium temperatures, consistent with the behavior expected for this ion displacement hole burning mechanism. While the spectral holes broaden with increased temperatures, this broadening is again largely reversed upon re-cooling from temperatures up to 70 K and a degree of persistence is observable to 100K. This material can provide a frequency reference for laser stabilization to the sub-KHz level.

A similar classification of the observed Er³⁺ centers into two families was found as for CaF₂:Tm³⁺:D⁻.

Gated spectral hole burning

Photon gated hole burning is also under investigation in a variety of compounds including the CaF₂ materials discussed above, a number of photorefractive materials, and materials with two-step photoionization hole burning mechanisms. These studies are on going.

Laser frequency stabilization using regenerative spectral hole burning Phys. Rev. B 62, 1473-1476 (2000)

We introduced and demonstrated a new laser frequency stabilization technique using *continuously* regenerated transient spectral holes. Regenerative hole widths to < 100 Hz provide extreme degrees of stabilization for time scales appropriate for many applications including spectroscopy, signal processing, ranging, and interferometry.

Stabilization to 20 Hz on a 10-ms time scale has been demonstrated using spectral holes at 793 nm in Tm³⁺:Y₃Al₅O₁₂ (Tm³⁺:YAG). Our experiments showed that this technique gives dramatic improvement in the reliability of stimulated photon echoes in the same spectral hole burning material. These lasers have also enabled the observation of a third population storage mechanism for hole burning in Tm³⁺:YAG.

This level of frequency stabilization of lasers is important for high-resolution spectroscopy, applications utilizing phase-sensitive detection such as precision laser ranging, long-baseline interferometry for gravitational wave detection, coherent optical communications, and time-domain spectral hole burning devices for functions such as: optical signal processing and packet switching and radio-frequency spectrum analysis. Modern frequency stabilization techniques use atomic or molecular resonances, such as iodine lines or laser-trapped ions or reflection modes of high-finesse Fabry-Perot interferometers, where cryogenic cavities can reduce frequency drift.

The new transient spectral hole frequency references are a complementary method to that described above for persistent spectral holes. A continuously-regenerated transient spectral hole combined with appropriate opto-electronic feedback acts as a viscous damping mechanism to restrict a laser's short term frequency variation. This is a fundamentally different procedure from locking to a fixed atomic transition, cavity resonance, persistent spectral hole, or instantaneous samples of phase or frequency history with delayed self-heterodyne detection.

Somewhat counter-intuitively, transient spectral holes in an inhomogeneously broadened absorption line do provide a useful reference for stabilizing a continuous wave laser. The stability is derived from the cumulative frequency memory of the hole, lasting over the hole lifetime and potentially longer in a well-engineered closed-loop feedback system where the regenerated holes

are exact enough copies. Other frequency reference techniques can provide greater precision on long time scales, but regenerative spectral hole burning provides both vibrational immunity and excellent short term stability, which are important and sufficient requirements for many applications beyond the field of spectral hole burning (SHB). Moreover, it should be possible to engineer a system of the type we report to fit into a compact shoebox-sized apparatus that includes optics, feedback electronics, and a cryostat.

An unstabilized laser initially burns a jitter-broadened spectral hole, the laser frequency immediately stabilizes to a fraction of that hole width, and the hole rapidly narrows to a limit set by the homogeneous linewidth of the material and the laser irradiance. This self-narrowing property of regenerated holes turns hole relaxation into an advantage. The balance between spontaneous hole decay and further hole burning from continued illumination determines the equilibrium depth of the regenerating hole.

Transient spectral hole burning may be achieved by a number of storage mechanisms. The most common is population storage in the excited state of an optically active ion or molecule, providing lifetimes of up to several tens of milli-seconds. Population storage in hyperfine components of the ground state can provide far longer lifetimes, up to 20 days. Far more systems exhibit transient SHB than exhibit persistent SHB, and this new stabilization strategy therefore gives access to a correspondingly greater range of frequencies. For the special case of SHB device applications that require a stable laser, transient spectral holes in an identical piece of the material used for the device naturally provide suitable stabilization at the needed wavelength.

We demonstrated this stabilization technique using the ${}^3H_6 \rightarrow {}^3H_4$ transition in Tm³⁺: Y₃Al₅O₁₂. at 793 nm. The fluorescence decay time of the upper state for these samples was measured to be 620 ms at 1.9 K, consistent with previous values. Decay to the intermediate 3F_4 state with a much longer fluorescence lifetime of around 12 ms occurs with a branching ratio of 0.54 and greatly lengthens the SHB storage time of this system.

The Tm³⁺:YAG crystals were grown by Scientific Materials Corp of Bozeman, MT. The Tm³⁺ concentrations of 0.1 at. % gave a peak absorption coefficient of 1.5 cm⁻¹ for the ${}^3H_6(1) \rightarrow {}^3H_4(1)$ transition, and an inhomogeneous linewidth of 20 GHz. Crystal thicknesses were 5.1 and 5.3 mm. The homogeneous linewidth of the transition is determined from the Mims dephasing time T_M of a two-pulse photon echo, determined experimental to be 75 ms in the absence of an applied magnetic field. In our more dilute samples we have measured T_M to be 116 ms, corresponding to an estimated full width at half maximum homogeneous linewidth of $\Gamma_h = 1/\pi T_M = 4$ kHz.

Using the apparatus of Fig. 1, above, two GaAlAs external cavity diode lasers were independently frequency stabilized to transient spectral holes at 793 nm in separate Tm³⁺:YAG crystals. A single cryostat held both crystals immersed in superfluid helium at 1.9 K. External phase modulation at 23 and 25 MHz, respectively, produced frequency side-bands with a modulation index of 0.22. The relative stability of the two lasers was measured by heterodyne detection of unmodulated portions of the beams. Beam diameters were approximately 1 mm, with an irradiance of 2.3 mW/cm².

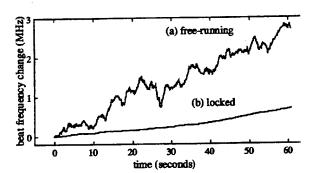


FIG. 5. Evolution of heterodyne beat signal between two lasers (a) free running, and (b) independently locked to transient spectral holes in the ${}^{3}H_{6} \rightarrow {}^{3}H_{4}$ transition in separate Tm³⁺:YAG crystals.

Evolution of the heterodyne beat frequency is shown in Fig. 5 for cases when both lasers were free running or both locked. The submegahertz free-running stabilities of these lasers are already sufficient for some spectroscopic applications, but stability was spectacularly improved by locking each laser to a transient spectral hole. On the time scale of seconds, a clear improvement has been made, although there is still a drift of about 10 kHz/s in this implementation. It is on time scales faster than this that the most significant stabilization occurs, as shown by the smoothness of the curve in Fig. 5(b). A quantitative measure of frequency stability on

specific time scales is provided by the two-sample or Allan variance. The root Allan variance is shown in Fig. 6. Minimum Allan variances of 20 Hz occur for time scales of 5-10 ms, representing more than three orders of magnitude improvement in stability over the free-running lasers.

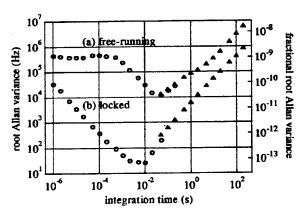


FIG. 6. Root Allan variance for the heterodyne beat frequency between two lasers (a) free running and (b) independently locked to transient spectral holes in the ${}^{3}H_{6} \rightarrow {}^{3}H_{4}$ transition in Tm³⁺:YAG. Circles are measured by a frequency counter, and triangles are computed from the data of Fig.

We believe that the major sources of instability are (a) residual amplitude modulation of the optical beam which causes voltage offsets upon mixing down to lower servo frequencies 20 and (b) thermally induced offsets and drift in the locking circuitry. These offsets corrupt the error signal and cause the laser frequency to lock slightly off the center of the hole, inducing drift as burning occurs at the shifted lock frequency. The present servo amplifier was adjusted to passively null the offset voltage at the start, but later fluctuations were uncompensated. The drift rate varied and changed directions on time scales of minutes indicating sensitivity to environmental changes. The frequency stabilization reported here was obtained without temperature stabilizing the electronics or optical setup and with vibration isolation provided by a standard pneumatically

floated optical table. The current 20 Hz stabilization with an 8 kHz resonance is not limited by any material properties, nor do there appear to be fundamental obstacles to reaching millihertz levels if the above sensitivities and offsets are reduced.

Statistically different noise sources have characteristic slopes on Allan variance plots. Broadband phase and frequency noise (slope = -1) limiting the left side of Fig. 6(b) may be reduced by selecting a quieter laser, a narrower spectral hole reference, or by increasing the fidelity and gain

bandwidth of the servo system. Frequency drift (slope = + 1) limits the right side of Fig. 6(b) and remains the primary barrier to attaining lower variances on time scales longer than 10 ms. Reduction of external vibrational and thermal influences may improve performance, however random frequency walks (slope = + 0.5) are not presently dominant factors on the time scales of Fig. 6.

With the high level of frequency stabilization achieved on millisecond time scales, this stabilization strategy provides ideal laser sources for optical coherent transient phenomena, in particular the photon echo and stimulated photon echo that are the basis for time-domain spectroscopy and optical devices. For optimal exploitation of the stimulated photon echo, laser frequency stability is required for the storage time of the material. Since this storage time is the lifetime of a transient spectral hole for the transition being probed, the requirement is naturally met by locking to a spectral hole.

Application of this frequency stabilization strategy and its potential in optical devices was demonstrated by measuring stimulated photon echoes on the ${}^3H_6 \rightarrow {}^3H_4$ transition of Tm³⁺:YAG using a frequency-stabilized laser. Approximately 1 mW of unmodulated continuous-wave power was available for producing echo excitation pulses after a portion of the laser output was phase modulated and used to frequency lock the laser to a regenerative transient spectral hole. The pulses were produced by two acousto-optic modulators, used in series to improve the on/off contrast ratio, with a third used after the crystal to block the excitation pulses. The photon echoes were detected with a thermoelectrically cooled C31034 photomultiplier. Three 1.5 μ s excitation pulses were incident on the sample, with the delay t_{12} between the first and second pulses fixed at 6 μ s. The stimulated photon echo was measured as a function of the delay t_{23} between the second and third pulses.

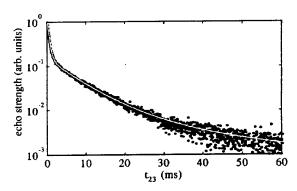


FIG. 7. Stimulated photon-echo decay on the ${}^{3}H_{6} \rightarrow {}^{3}H_{4}$ transition in Tm³⁺:YAG, measured with a laser stabilized to a transient spectral hole, showing three distinct population storage mechanisms. Each point represents a single-shot event.

With the laser frequency locked to a transient spectral hole, photon echoes could be measured consistently for t_{23} delay times of several tens of milliseconds, giving the data in Fig. 7. The limiting factor for measuring echoes with longer t_{23} delay times was the signal-to-noise ratio, rather than laser frequency jitter. In contrast, when the stimulated echo decay was measured with the laser free running, the reproducibility of the stimulated echo was unreliable after only 500 µs, as shown in Fig. 8. The data points of Fig. 7 and Fig. 8 were single-shot acquisitions of the stimulated photon echo without thresholding to reject low-intensity echoes. In Fig. 8 it was clear that frequency jitter was the cause of the echo signal amplitude fluctuations, since occasionally a true-valued echo was

produced when the laser frequency of the third pulse happened to match that of the first two. An envelope of true-valued echoes can be seen, but most points fall well below this. Clearly, averaging the data in Fig. 8 over multiple shots would lead to a much different and erroneous echo decay rate.

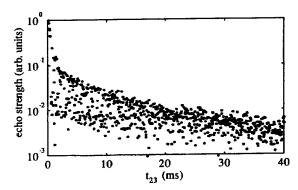


FIG. 8. Stimulated photon-echo decay on the ${}^{3}H_{6} \rightarrow {}^{3}H_{4}$ transition of Tm³⁺:YAG, without frequency stabilization of the laser. Each point represents a single-shot event.

The generation of a stimulated photon echo can be considered as the scattering of the excitation pulse off the population grating generated by the first two pulses. The first two pulses create a modulation in the population of the excited state as a function of frequency, and a corresponding depletion in the ground state. The electric-field vector of the echo stimulated by the excitation pulse is proportional to the sum of these two gratings as they exist at the time of the excitation pulse. Intermediate state populations do not contribute directly to the echo but allow the ground-state depletion to remain for longer than the lifetime of the upper state. A rate equation analysis for a four-level

system shows that the echo electric field decays with increasing t_{23} delay time as the sum of three exponential functions whose decay times are the lifetimes of the three excited states involved. The detected echo strength is the square of this function.

The square root of the echo intensity was fitted to three exponentials giving the solid white line in Fig. 7 with decay times of 590 μ s, 11.8 ms, and 90 ms. The first decay time corresponds to population storage in the 3H_4 excited state of the transition, in agreement with the 620 μ s value obtained from fluorescence decay. The second corresponds to population storage in the intermediate 3F_4 metastable state, a mechanism previously shown to account for transient spectral hole burning in Tm^{3+} doped crystals. The third, longest decay component has an uncertainty of about 50% for its decay time due to the scatter of the data attributed to detector noise. This component is assigned to an energy shift arising from the coupling of Tm^{3+} to the nuclear spins of lattice Al^{3+} ions. To confirm the nuclear-spin coupling contribution to the population storage, the stimulated echo decay was remeasured with a permanent magnet placed immediately beneath the cryostat. This produced a very modest magnetic field at the crystal (~100G), but it was enough to show a distinct increase of the decay time associated with this level structure, corresponding to increased spin-lattice relaxation times.

Regenerative transient spectral hole burning can provide an effective means for stabilizing the frequency of a laser. A high degree of stabilization can be achieved, as the spectral holes in some materials can be narrower than 100 Hz. This stabilization method is well suited for spectroscopy and for optical data processing devices based on time-domain spectral hole burning since separate pieces of the same material can be used as stabilizer and processor. A substantial improvement in stimulated photon-echo reproducibility was demonstrated.

More elaborate signal processing experiments by other groups at MSU show that the superiority of these stabilized lasers is truly *enabling* in terms of application of spectral hole burning signal processors.

Laser Stabilization at 1536 nm Using Regenerative Spectral Hole Burning To be submitted to Phys. Rev. B

Laser frequency stabilization to 500 Hz on a 2 ms time scale with drift reduced to 7 kHz/min over several-minutes was demonstrated in the optical communication band at 1536 nm. A continuously-regenerated spectral hole in the inhomogeneously broadened $^4I_{15/2}(1) \rightarrow ^4I_{13/2}(1)$ optical absorption of an Er^{3+} :Y₂SiO₅ crystal was used as the short term frequency reference, while a variation on the locking technique allowed simultaneous use of the inhomogeneously broadened absorption line as a long term reference. The reported frequency stability was achieved without vibration isolation. Spectral hole burning frequency stabilization provides ideal laser sources for high resolution spectroscopy, real time optical signal processing, and a range of applications requiring ultra-narrowband light sources or coherent detection; the time scale for stability and the compatibility with spectral hole burning devices make this technique complementary to other frequency references for laser stabilization.

The availability of ultra-narrow SHB resonances down to 15 Hz in rare earth doped crystals, the relative immunity of spectral holes to environmental disturbances such as vibrations, and the portability and compactness of a stable laser system using SHB references with a closed cycle cryocooler are important features that should enable application in a variety of fields beyond those normally associated with spectral hole burning. Stabilization of mode locked lasers to spectral holes also should be practical and will have applications in the signal processing situations discussed above and in other contexts that require short pulses, frequency combs, or optical clocks. The SHB frequency references are well suited to applications where multiple frequencies are required and where the programmability of SHB materials allows programmable frequency differences up to the multi-GHz range or, if disordered solids are used, to the THz range. With the development of suitable photon-gated (or two-photon) SHB materials, the production of long term secondary frequency standards based on SHB may become practical.

When stabilized laser sources are required for real time optical signal processing in SHB materials, the use of a second piece of the same signal processing material as a SHB frequency reference provides automatic frequency compatibility between the signal processing material and the stabilized laser source. The relative vibrational immunity of the spectral holes provides an important simplification in system design and performance for either spectroscopy or SHB devices; this advantage is even greater when both the frequency reference and spectroscopic sample or SHB device are mounted on the same platform or sample holder. This has been demonstrated here for Er^{3+} :Y₂SiO₅. A stabilized laser of this type would be especially helpful, for example, in measurements of spectral diffusion using the stimulated photon echo technique.

The importance of stabilization in SHB signal processing is underscored by the observation that early moderate-speed demonstrations have been limited by laser frequency jitter that led to a loss in signal fidelity. These problems can occur at several levels: a) uncontrolled phase variations between programming pulses when repeated pulse sequences are used for writing or refreshing spectral interference gratings, b) the more extreme case where the jitter exceeds the Fourier width of the exciting pulses so that the processed pulses fail to overlap spectrally with the programming pulses, and c) the case where the jitter exceeds the Fourier width of the exciting pulses so that the excitation pulses fail to overlap spectrally with the probe pulse in measurements of spectral diffusion. Lasers stabilized to spectral holes are already playing an important role in proof-of-principle demonstrations of a variety of SHB devices.

An Er³⁺-doped SHB crystal was used to demonstrate laser stabilization in the important 1.5 µm telecommunications band. Studies have shown that Er³⁺-doped crystals have the frequency selectivity required for optical storage, real-time address header decoding for all-optical packet routing, and all-optical correlation. Since the laser in the present report is stabilized to the same transition used in those device demonstrations, the requirements of frequency overlap, frequency and phase stability, and time scale of stabilization are all automatically fulfilled, as mentioned earlier. The limits on device performance are set then by material parameters rather than by instability of the laser.

The stabilized diode laser - Er³+:Y₂SiO₅ system described here exploits our regenerative SHB technique described in a previous section. A transient spectral hole is continuously regenerated by the stabilized laser and provides a frequency reference at an arbitrarily chosen location in the inhomogeneous Er³+:Y₂SiO₅ absorption profile. The stability of the laser will then be determined by the dynamical properties of the SHB material together with the design of the locking system.

The SHB crystal chosen as a frequency reference was Er3+: Y2SiO5 with an Er3+ concentration of 0.005 atomic percent. This material exhibits transient spectral hole burning on the ${}^{4}I_{15/2}(1) \rightarrow {}^{4}I_{13/2}(1)$ transitions at 1536.14 nm (site 1) and 1538.57 nm (site 2) by population storage in the excited state of the optically active ion. The inhomogeneous linewidth $\Gamma_{inh} = 500 \text{ MHz} = 0.017 \text{ cm}^{-1}$. The homogeneous linewidth Γ_h of this Er³⁺ transition decreases substantially in applied magnetic fields, leading to narrower spectral hole widths. The homogeneous linewidth for this crystal was determined from the optical dephasing time T2 obtained from two-pulse photon echo decay experiments at 1.6 K, and the measured value $\Gamma_h \sim 5$ kHz is consistent with previously published results. Stronger magnetic fields have reduced. the homogeneous line width to 78 Hz, and it is expected that further elucidation of the angular dependence of the Zeeman splittings will reveal a field direction that provides more rapid freezing out of electronic spin flips involving the excited component of the ground state and thus allow the linewidth to more closely approach the T₁ lifetime limited value of 15 Hz. The minimum line width of a shallow spectral hole burned by a narrow-band laser is 2 Γ_h due to convolution of burning and reading cycles; deeper holes become broader, since there is less saturation of material absorption in the wings of a hole than at the center.

The frequency locking experiments reported here were performed using the strongest absorption transition from the lowest Zeeman-split level for site 1 in moderate magnetic fields of B=0.2 to 0.5 T; the 0.2 T magnetic field value was chosen to simulate field strengths that have been obtained using compact 1 cm dia. Nd-Fe-B permanent magnets. The homogeneous linewidth at 0.5 T is about half that at 0.2 T.

The experimental apparatus was similar to that shown in Fig. 1. Two shop built external cavity diode lasers in the Littman-Metcalf configuration were equipped with InGaAsP/InP quantum well diodes that had one facet angled to eliminate intra-cavity optical feedback. The Pound-Drever-Hall technique was used for locking the laser frequency to the spectral hole. The error signal was derived from the spectral hole transmission using frequency modulation (FM) spectroscopy, with the two lasers modulated by external electro-optic modulators driven at 27 MHz and 30 MHz, respectively. These frequencies greatly exceeded the spectral hole widths but were far less than the inhomogeneous absorption linewidth $\Gamma_{\rm inh} = 500$ MHz. The primary laser side bands had a modulation index M = 0.4, and secondary side bands were small but observable. Time-domain

spectroscopy and a wide range of proposed SHB optical devices are based on the photon echo and stimulated photon echo, the capabilities of these techniques can be improved with the level of frequency stabilization reported here. For optimal exploitation of the stimulated photon echo, laser frequency stability to better than the spectral width of the broadest excitation pulse, or in the limiting case to better than a homogeneous linewidth, is required for the storage time of the material, which is defined by the decay time of a transient spectral hole for the transition being probed. With lasers stabilized to spectral holes, this requirement is naturally and automatically met.

With the laser frequency locked to a transient spectral hole. photon echoes could be measured consistently for t_{23} delay times of several hundreds of microseconds. The limiting factor for measuring echoes with longer t_{23} delay times was the detector signal-to-noise ratio, rather than laser frequency jitter, even though additional jitter may have been introduced by an erbium-doped fiber amplifier (EDFA) used in the echo measurements. After 800 μ s total delay time the stimulated echo signal was buried in the noise. In contrast, when the stimulated echo decay was measured with the laser free running, the reproducibility of the stimulated echo became unreliable after only 200 μ s. As for the Tm³+: YAG case described earlier, an envelope of "good" echoes can be seen, but most points fall well below this. Clearly, averaging the data over multiple shots would lead to a much different and erroneous echo decay rate. This not only shows the potential for improving spectroscopic measurements with our stabilized lasers, but it also demonstrates their impact on signal to noise and reproducibility in optical signal processing in the communications band with spectral hole burning materials.

Laser Development

We constructed four external cavity diode lasers for this project. Two are tunable from 792 nm to 814 nm and may be continuously scanned with single-mode output over 20 GHz ranges. This tuning and scanning range was adequate to cover the Tm³+-based materials of interest for stabilization in this spectral range. These lasers make excellent projects for undergraduate students. Design and construction of our own lasers also gives us the opportunity to explore and have an extra degree of control over parameters that are important for these stable laser applications.

Two of the lasers are tunable over the 1.5 micron communications band to cover the Er³⁺-based materials of interest for stabilization in this spectral range. The use of single angled facet diodes has been explored for this wavelength region.

Our diode lasers were first locked to the optical resonances of a Fabry-Perot cavity that has been constructed for the initial phase locking experiments. This was used for basic optimization of the electronic feedback stabilization circuits and optical systems. The inputs for scanning the laser frequency are angle tuning of the diffracted optical feedback by a piezoelectric transducer at 240 MHz/V plus diode temperature tuning and current tuning at 12 GHz/C.

Rapid success with locking to spectral holes in early stages of the project demonstrated that locking to cavities was not a necessary step in achieving stabilization to spectral holes. Locking to cavities remains a useful diagnostic stage in setting up new lasers or systems, but it is not a required step in operation of our stable laser systems.

Personnel Supported and Associated

Faculty Principal Investigators were Prof. Rufus L. Cone and Prof. John L. Carlsten.

Two postdocs worked on the project full time for the duration of the project.

- Dr. Nicholas M. Strickland joined the group on November 3, 1996, from the Department of Physics and Astronomy, University of Canterbury, Christ Church New Zealand. His Ph. D. Thesis with Dr. Glynn Jones, was entitled, "Laser Excitation and Infrared Absorption Spectroscopy of Rare-Earth Ion Centres in Fluorite Crystals." Dr. Strickland left the group in September, 1999, at the end of the project.
- Dr. Peter B. Sellin joined the group in December, 1996, from the Physics Department of the University of Oregon, Eugene, Oregon. His Ph. D. Thesis with Prof. Thomas W. Mossberg was entitled, "Inversionless Gain, Irreversible Mechanisms, and Field-Assisted Lasing in Driven Atoms." Dr. Sellin left the group in September, 1999, at the end of the project.

A summer undergraduate, Zane Aldworth, from the University of Puget Sound, Tacoma, Washington, was supported by the Montana Space Grant Consortium to work full time on this project in the summer of 1997.

Several MSU undergraduates including Mechanical Engineering student Casey Dodge and Physics students Glenn Omdahl, Seth Meyer, Kevin Yager, Malina Schindel, Andrew Schmidt, and Anna Hagenston worked on the project under support of an AASERT grant.

Publications Produced By This Project

- 1. Programmable frequency reference for sub-kHz laser stabilization using persistent spectral hole burning, P. B. Sellin, N. M. Strickland, J. L. Carlsten, and R. L. Cone, Opt. Lett. 24, 1038-1040 (1999).
- 2. Persistent Spectral Hole Burning in Deuterated CaF₂: Tm³⁺, N. M. Strickland, R. L. Cone, and R. M. Macfarlane, Phys. Rev. B. **59**, 14328-14335 (1999).
- 3. Laser Frequency Stabilization using Regenerative Spectral Hole Burning, N. M. Strickland, P. B. Sellin, Y. Sun, J. L. Carlsten, and R. L. Cone, Phys. Rev. B 62, 1473-1476 (2000).
- Laser Stabilization at 1536 nm Using Regenerative Spectral Hole Burning, P. B. Sellin, N. M. Strickland, T. Böttger, J. L. Carlsten, and R. L. Cone, submitted to Phys. Rev. B.
- 5. Persistent Spectral Hole Burning in Deuterated CaF₂:Er³⁺, N. M. Strickland, R. L. Cone, and R. M. Macfarlane, for submission to Phys. Rev. B.

Interactions

Presentations and Participation at Meetings and Conferences

Invited Conference Talk, *Materials for Coherent Transient Optical Memories and Signal Processors*, R. L. Cone, 5th International Meeting on Hole Burning and Related Spectroscopies, Brainerd, MN September 13-17, 1996.

Invited Talk, Materials and Material Concepts for Devices at Infrared, Visible, and Communication Wavelengths Based on Ions in Crystals, R. L. Cone, Workshop on Applications of Persistent Spectral Hole Burning, Big Sky, MT, February 16-19, 1997.

Invited Talk, Ultranarrow Lines in Rare Earth Crystals and Frequency References Based on Persistent Spectral Hole Burning, R. L. Cone, Texas A&M Quantum Optics Workshop on Lasing Without Inversion, Taos, NM, August 14-16, 1997.

Refereed Conference Talk, Measurement of Photon Echoes at 1.5 microns in Er^{3+} : Y_2SiO_5 using a Diode Laser and Amplifier, R. M. Macfarlane, T. L. Harris, Y. Sun, R. L. Cone, and R. W. Equall, refereed summary accepted for presentation at the Quantum Electronics and Laser Science Conference (CLEO/QELS), Baltimore, MD, May, 1997.

Invited Talk, Material Design and Characterization for Hole Burning Devices and Optical Frequency Standards, R. L. Cone, Optical Science and Laser Technology Conference, Montana State University, Bozeman, MT, August 25-26, 1997.

Invited Talk, Diode Laser Control, Characterization, and Conversion, J. L. Carlsten, Optical Science and Laser Technology Conference, Montana State University, Bozeman, MT, August 25-26, 1997.

Contributed Talk, Erbium based materials for data routing and processing at 1550 nm, Y. Sun, T. L. Harris, R. L. Cone, R. M. Macfarlane, and R. W. Equall, Optical Science and Laser Technology Conference, Montana State University, Bozeman, MT, August 25-26, 1997.

Contributed Talk, Persistent Spectral Hole Burning in Rare Earth Doped Deuterated Calcium Fluoride, N. Strickland, R. L. Cone, R. M. Macfarlane, Optical Science and Laser Technology Conference, Montana State University, Bozeman, MT, August 25-26, 1997.

Contributed Talk, Ultra-Stabilized Diode Lasers and Persistent Spectral Holes: Developing a Compact and Tunable Frequency Standard, P. B. Sellin, N. M. Strickland, Z. N. Aldworth, J. L. Carlsten, and R. L. Cone, Optical Science and Laser Technology Conference, Montana State University, Bozeman, MT, August 25-26, 1997.

Invited Talk, New Rare Earth Spectral Hole Burning Materials, Material Characterization, and Applications," Fourth International Workshop on Applications of Spectral Hole Burning, Montana State University, Bozeman, MT, March 9-11, 1998.

Frequency Locking of Diode Lasers, and Frequency References, P. B. Sellin, J. L. Carlsten, N. M. Strickland and R. L. Cone, Fourth International Workshop on Applications of Spectral Hole Burning, Montana State University, Bozeman, MT, March 9-11, 1998.

Er³⁺ Materials for All-Optical Switching and Routing at 1.5 μm, Y. Sun, R. L. Cone, T. L. Harris, R. M. Macfarlane, and R. W. Equall, March Meeting of the American Physical Society, Los Angeles, CA, March 15 - 20, 1998.

Tm³⁺ Materials for Coherent Transient Signal Processing Near 790 nm, R. L. Cone, G. M. Wang, Y. Sun, R. M. Macfarlane, March Meeting of the American Physical Society, Los Angeles, CA, March 15 - 20, 1998.

Persistent Spectral Hole Burning in CaF₂: Tm³⁺:D, N. M. Strickland, R. L. Cone, Physics Department, R. M. Macfarlane, March Meeting of the American Physical Society, Los Angeles, CA, March 15 - 20, 1998.

Session Chair, M26 IMSTG: Near Field Microscopes – Optical, March Meeting of the American Physical Society, Los Angeles, CA, March 15 - 20, 1998.

Session Chair, O15 DCMP: Optical Properties -- Ions in Solids, March Meeting of the American Physical Society, Los Angeles, CA, March 15 - 20, 1998.

Invited Conference Talk, Advances in Rare Earth Crystals for Spectral Hole Burning Applications and Frequency References, R. L. Cone, Texas A&M – Office of Naval Research Quantum Optics Workshop, Jackson, Wyoming, August 2-6, 1998.

Advances in Rare Earth Crystals for Coherent Transient Applications and Lasers, R. L. Cone, Optical Science and Laser Technology Conference, Montana State University, Bozeman, MT, Aug. 17 - 18, 1998.

Er³⁺ Materials for All-Optical Switching and Routing at 1.5 μm, Y. Sun, T. L. Harris, and R. L. Cone, R. M. Macfarlane, R. W. Equall and R. L. Hutcheson, Optical Science and Laser Technology Conference, Montana State University, Bozeman, MT, Aug. 17 - 18, 1998.

Frequency Stabilization of External Cavity Diode Lasers, P. B. Sellin, N. M. Strickland, J. L. Carlsten, and R. L. Cone, Optical Science and Laser Technology Conference, Montana State University, Eozeman, MT, Aug. 17 - 18, 1998.

Invited Talk, New Rare Earth Materials, Material Concepts, and Prototype Devices, R. L. Cone, Y. Sun, F. Könz, N. M. Strickland, T. L. Harris, C.W. Thiel, T. Böttger, G. Reinemer, and W. R. Babbitt, R. W. Equall and R. L. Hutcheson, R. M. Macfarlane, J. A. Ritcey, Fifth International Workshop on Applications of Spectral Hole Burning, Montana State University, Bozeman, MT, March 7-10, 1999.

Laser Frequency Stabilization to the kHz Level using Persistent Spectral Hole Burning, P. B. Sellin, N. M. Strickland, J. L. Carlsten and R. L. Cone, Fifth International Workshop on Applications of Spectral Hole Burning, Montana State University, Bozeman, MT, March 7-10, 1999.

Frequency Stabilization of an External Cavity Diode Laser Operating at 1550nm for Spectral Hole Burning Applications, T. Böttger, T. L. Harris, G. C. Dodge, P. B. Sellin, and R. L. Cone, Fifth International Workshop on Applications of Spectral Hole Burning, Montana State University, Bozeman, MT, March 7-10, 1999.

Programmable Frequency Reference for Laser Stabilization Using Persistent Spectral Hole Burning, P. B. Sellin, N. M. Strickland, J. L. Carlsten and R. L. Cone, Conference on Lasers and Electro-Optics (CLEO), Baltimore, MD, May 23 - 28, 1999.

INVITED PLENARY CONFERENCE TALK, Applications of Spectral Hole Burning, R. L. Cone, First Meeting of the Northwest Section of the American Physical Society, Vancouver, BC, Canada, May 21 - 22, 1999.

Programmable Frequency Reference For Laser Stabilization Using Persistent Spectral Hole Burning, P. B. Sellin, N. M. Strickland, J. L. Carlsten, and R. L. Cone, Conference on Lasers and Electro-Optics (CLEO), Baltimore, MD, May 23 - 28, 1999.

Invited Conference Talk, Photon Echo Characterization of Er³⁺ Doped Oxide Materials at 1.5 µm, Y. Sun, R. L. Cone, T. L. Harris, R. M. Macfarlane, and R. W. Equall, 12th International Conference on Dynamical Processes in Excited States of Solids, Humacao, Puerto Rico, May 23 - 27, 1999.

Invited Conference Talk, *Photon Echoes and Signal Processing Applications of Erbium Doped Oxides*, Y. Sun, R. L. Cone, T. L. Harris, R. W. Equall, R. L. Hutcheson, R. M. Macfarlane, 22nd Rare Earth Research Conference, Argonne National Laboratory, Advanced Photon Source Conference Center, Argonne, IL, July 10 - 15, 1999.

Invited Conference Talk, Rare Earth Spectroscopy and Laser Frequency Stabilization to Ultranarrow Spectral Holes, R. L. Cone, Texas A&M – Office of Naval Research Quantum Optics Workshop, Jackson, Wyoming, July 26 - 30, 1999.

Invited Talk, Advances in Hole Burning Materials, Optical Frequency Standards, and Lasers for Remote Sensing, R. L. Cone and Y. Sun, 1999 Optical Science and Laser Technology Conference, Montana State University, Bozeman, Montana, August 16-17, 1999.

An Optical Shock Absorber: Laser Frequency Stabilization to 20 Hz using Regenerative Spectral Hole Burning, N. M. Strickland, P. B. Sellin, Y. Sun, J. L. Carlsten, R. L. Cone, 1999 Optical Science and Laser Technology Conference, Montana State University, Bozeman, Montana, August 16-17, 1999.

Programmable Frequency Reference for Sub-kilohertz Laser Stabilization by Persistent Spectral Hole Burning, P. B. Sellin, N. M. Strickland, J. L. Carlsten and R. L. Cone, 1999 Optical Science and Laser Technology Conference, Montana State University, Bozeman, Montana, August 16-17, 1999.

Concentration, Temperature, and Composition Dependence of Optical Dephasing and Absorption in Eu³⁺:Y₂SiO₅ for Optical Memory and Frequency Standards, F. Könz, Y. Sun, C. W. Thiel, R. L. Cone, R. W. Equall, R. L. Hutcheson, and R. M. Macfarlane, 1999 Optical Science and Laser Technology Conference, Montana State University, Bozeman, Montana, August 16-17, 1999.

Progress in Photoemission and Photoconductivity Measurements on Hole Burning Materials and Laser Materials, C. W. Thiel, Y. Sun, R. L. Cone, Herve Cruguel, G. J. Lapeyre, R. W. Equall, and R. M. Macfarlane, 1999 Optical Science and Laser Technology Conference, Montana State University, Bozeman, Montana, August 16-17, 1999.

Frequency Stabilization of an External Cavity Diode Laser Operating at 1.5 µm for Spectral Hole Burning Applications, T. Böttger, T. L. Harris, G. C. Dodge, P. B. Sellin, N. M. Strickland, and R. L. Cone, 1999 Optical Science and Laser Technology Conference, Montana State University, Bozeman, Montana, August 16-17, 1999.

Invited Talk, Rare Earth Spectral Hole Burning Materials, R. L. Cone, 1st Lyon Workshop on Nano-Optics, Universite Claude Bernard, Lyon, France, September 15 and 16, 1999.

Program Committee, 6th International Conference on Hole-Burning and Related Spectroscopies, Hourtin, France, September 18 - 23, 1999

Invited Conference Talk, New Rare Earth Materials, Material Concepts, and Demonstrations of Spectral Hole Burning Applications, R. L. Cone, Y. Sun, F. Könz, N. M. Strickland, T. L. Harris, C. W. Thiel, T. Böttger, G. Reinemer, W. R. Babbitt, R. W. Equall, R. L. Hutcheson, R. M. Macfarlane, and J. A. Ritcey, 6th International Conference on Hole-Burning and Related Spectroscopies, Hourtin, France, September 18 - 23, 1999.

Concentration and Temperature Dependence of Optical Dephasing and Hyperfine Spectral Hole Relaxation in Eu³⁺:Y₂SiO₅, F. Könz, Y. Sun, C. W. Thiel, R. L. Cone, R. W. Equall, and R. L. Hutcheson, 6th International Conference on Hole-Burning and Related Spectroscopies, Hourtin, France, September 18-23, 1999.

Persistent spectral hole burning in deuterated CaF₂: Tm³⁺ and CaF₂: Er³⁺ and application to laser frequency stabilization, N. M. Strickland, P. B. Sellin, J. L. Carlsten, R. L. Cone, and R. M. Macfarlane, 6th International Conference on Hole-Burning and Related Spectroscopies, Hourtin, France, September 18 - 23, 1999.

Invited Talk, Spectral Hole Burning for Laser Spectroscopy and Applications, R. L. Cone, La Societe Française de Physique, Section Rhone-Loire, Universite Lyon I, Lyon, France, September 30, 1999.

Invited Talk, Lasers Stabilized to Spectral Holes and a Survey of Hole Burning Materials for Signal Processing, R. L. Cone, Laboratoire de Physico-Chemie Materiaux Luminescents, Universite Lyon I, Lyon, France, October 4, 1999.

Consultative and Advisory Functions

Cone's group served as advisors on crystal design and characterization to Scientific Materials Corporation of Bozeman, MT, an AFOSR SBIR Phase II contractor. A number of new materials were developed in this role. Cone's group also worked to enhance linkage of Scientific Materials to other groups in the Spectral Hole Burning community. Together Scientific Materials and Cone's group served a primary role as material developers and providers for application and advancement of spectral hole burning technology.

Carlsten's group served as advisors on laser optics technology to ILX Lightwave of Bozeman, MT.

These interactions included participation in yearly AFOSR Workshops:

 Third Workshop on Applications of Persistent Spectral Hole Burning, Big Sky, MT, February 16-19, 1997.

- Fourth International Workshop on Applications of Spectral Hole Burning, Montana State University, Bozeman, MT, March 9-11, 1998.
- Fifth International Workshop on Applications of Spectral Hole Burning, Montana State University, Bozeman, MT, March 7-10, 1999.

Professor John Carlsten, Peter B. Sellin, and Nicholas Strickland visited two of the leading laboratories in laser stabilization to establish contacts, to learn about their programs, and to see laboratory facilities.

They visited Prof. J. L. Hall at JILA in Boulder, Colorado, on December 10, 1996 and discussed:

- Laser stabilization of dye, He-Ne, and diode lasers,
- Amplifiers, filters, feedback circuitry,
- Phase noise stabilization in optical fibers,
- Frequency standard chains,
- Vibration isolation,
- Electro-optic modulator design and construction.

They visited L. Hollberg at NIST in Boulder, Colorado, on December 11, 1996 and discussed:

- Diode laser stabilization,
- Laser trapped atomic calcium frequency standard,
- Diode Anti-reflection coating apparatus,
- Fast Extended Tuning of Diode Lasers.

Cone has corresponded with Stephan Schiller of the Quantum Metrology Group of the Universitat Konstanz, Germany, on narrow band lasers. Cone and Carlsten met with Schiller and discussed mutual interests at the 1997 Conference on Lasers and Electro-Optics (CLEO). Schiller shares our interest in Eu³⁺:Y₂SiO₅ and his group has produced unusual sources in the relevant 590 nm region.

Transitions:

The technology for stabilization to spectral holes has been transferred to several other research groups at MSU, leading to significant advancements in their AFOSR and DoD funded programs:

- Professor W. R. Babbitt's group on optical signal processing has exploited the technology in sophisticated demonstrations of correlators and true-time delay generators and RF spectrum analyzer project.
- Dr. Kris Merkel and Alan Craig have used a Ti:S laser stabilized to spectral holes to demonstrate the accumulation of complex correlator signals for optical signal analysis.

Groups in France have also expressed a need for these techniques in their correlator and RF spectrum analyzers.

Scientific Materials Corporation and others are interested in commercializing these lasers.

Two patents have been applied for.

- 1. Programmable Frequency Reference For Laser Frequency Stabilization, And Arbitrary Optical Clock Generator, Using Persistent Spectral Hole Burning

 This application is related to stabilizing the frequency of a tunable laser and, more particularly, to using spectral hole burning materials to stabilize the frequency of a tunable laser.
- 2. Laser Frequency Stabilizer Using Transient Spectral Hole Burning

 This application relates to a device whereby the frequency of a laser is actively stabilized by locking to a transient spectral hole in a solid state or other condensed phase reference material.